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(54) **TWO STAGE DRY FEED GASIFICATION SYSTEM AND PROCESS**

SYSTEM UND VERFAHREN FÜR ZWEISTUFIGE TROCKENSTOFFVERGASUNG

SYSTÈME ET PROCÉDÉ DE GAZÉIFICATION ALIMENTÉS EN MATIÈRES SÈCHES ET À DEUX ÉTAGES

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Description

FIELD OF THE DISCLOSURE

5 **[0001]** The present invention relates to a gasification system and process for converting generally solid feedstock such as carbonaceous material into desirable gaseous products such as synthesis gas.

BACKGROUND

10 **[0002]** Gasification processes are widely used to convert solid or liquid feedstocks such as coal, petroleum coke and petroleum residue into synthesis gas (syngas). Syngas is an important intermediate feedstock for producing chemicals such as hydrogen, methanol, ammonia, synthetic natural gas or synthetic transportation oil. Syngas can also be used to generate electricity through a process known as Integrated Gasification Combined Cycle (IGCC).

15 **[0003]** The common practice for gasification processes is to contact a feedstock with oxygen directly above the auto-ignition temperature of the fuel. The drawback of this practice is that a portion of the combustion heat is consumed to heat up the feedstock and to vaporize the moisture carried in by the feedstock, the end result being a reduction in the energy efficiency of the process. Lower energy efficiency translates to higher feedstock consumption and greenhouse gas emissions. Thus, there is a need to develop a gasification system that overcomes the above drawback.

20 **[0004]** US 4 069 024 A describes a two-stage coal gasification system in which high temperature synthesis gas produced in a downflowing gasifier containing a high quantity of sensible heat is subsequently reacted in a pyrolyzing reactor with a charge of carbonaceous material and limestone at a reduced temperature to utilize the sensible heat contained in the gas and simultaneously produce an increase in the hydrocarbon content thereof.

25 **[0005]** WO2008/138166 A1 describes a coal circulating fluidized bed coal gas generating furnace system comprising the following successively connected equipments: a coal gas generating furnace, a high temperature separator, a heat exchanger, a low temperature separator and a waste heat boiler.

SUMMARY

30 **[0006]** The present disclosure relates to a dry feed two stage gasification system and process for gasifying feedstock such as carbonaceous materials with improved energy efficiency along with lower feedstock consumption and CO₂ emissions.

35 **[0007]** Certain embodiments describe a process for gasifying a carbonaceous material comprising the steps of: (a) providing a gasification reactor comprising a reactor upper section and a reactor lower section; (b) introducing a substantially dry solid carbonaceous feedstock stream into the reactor upper section and reacting therein with a first mixture product from the reactor lower section, thereby forming a second mixture product, wherein the first mixture product comprises syngas, and wherein the second mixture product comprises a second solid product and a second gaseous product; (c) passing the second mixture product to a first separating device, wherein the second solid product is separated from the second gaseous product; (d) passing the separated second solid product into the reactor lower section; (e) passing the second gaseous product stream exiting the first separating device through a heat recovery unit, thereby
40 lowering the temperature of the second gaseous product stream to provide a cooled second gaseous product and creating steam; (f) passing the steam produced in step (e) into the reactor lower section; (g) pulverizing a solid carbonaceous feedstock; (h) mixing the pulverized solid carbonaceous feedstock with the cooled second gaseous product of step (e) by means of a solid-gas mixer to form a warm solid-gas mixture, wherein said warm solid-gas mixture is maintained at a temperature in a range of 148°C (300°F) to 537°C (1000°F); (i) separating the warm solid-gas mixture
45 in a second separating device (150) to produce the substantially dry solid carbonaceous feedstock of step (b) and a separated second gaseous product; (j) passing the separated second gaseous product exiting the second separating device to a particulate filtering device, wherein a stream of residual solids, fines and particulates is removed and passed into the reactor upper section; and (k) combining a gas stream, the separated second solid product stream of step (d) and the steam produced in the heat recovery unit of step (e) in the reactor lower section and reacting them, thereby
50 evolving heat and forming the first mixture product comprising syngas of step (b), wherein the gas stream comprises an oxygen supply selected from the group consisting of an oxygen-containing gas, steam, and mixtures thereof.

55 **[0008]** The substantially dry solid carbonaceous feedstock stream and the residual solids fines and particulates may be introduced to the reactor upper section by one or more feeding devices. The gas stream, the second solid product exiting the first separating device, and the steam produced from the heat recovery unit maybe introduced into the reactor lower section by one or more dispersion devices. The carbonaceous material is selected from the group consisting of coal, lignite, petroleum coke and mixtures thereof. The oxygen-containing gas may be selected from air, oxygen-enriched air, oxygen and mixtures thereof. The heat recovery unit may be selected from the group consisting of a radiant heat type boiler, water tube boiler, fire tube boiler and combinations thereof. The first and second separating devices may

each comprise a cyclone. The temperature of the second mixture product leaving the upper section of the gasifier may be between about 648°C (1200°F) and 1371°C (2500°F) prior to entering into the first separating device, but preferably is between 815°C (1500°F) and 1093°C (2000°F). The warm solid-gas mixture is preferably maintained at a temperature in the range of 260°C (500°F) to 426°C (800°F).

5 **[0009]** Certain embodiments relate to a system for gasifying a carbonaceous material including: (a) a reactor upper section for reacting a substantially dry solid carbonaceous feedstock, a stream of recycled residual solids, fines and particulates, and a first mixture product from a reactor lower section to produce a second mixture product, wherein the first mixture product comprises syngas, and the second mixture product comprises a second solid product stream and a second gaseous product stream; (b) a first separating device for separating the second solid product stream from the second gaseous product stream, wherein the second solid product stream is introduced into the reactor lower section; 10 (c) a heat recovery unit for cooling the temperature of the second gaseous product stream to provide a cooled second gaseous product and producing steam, wherein said steam is passed into the lower reactor section; (d) a solid-gas mixer (160) for mixing a pulverized solid carbonaceous feedstock with the cooled second gaseous product of part (c) to form a warm solid-gas mixture, wherein the solid-gas mixer (160) is configured for maintaining the warm solid-gas mixture at a temperature in a range of 148°C (300°F) to 537°C (1000°F); (e) a second separating device (150) for separating the warm solid-gas mixture to produce the substantially dry solid carbonaceous feedstock of part (a) and a separated second gaseous product; (f) a particulate filtering device for separating the residual solids, fines and particulates from the separated second gaseous product stream exiting the second separating device, wherein the separated residual solids, fines and particulates are passed into the reactor upper section; (g) a reactor lower section for reacting a mixture 20 comprising the separated second solid product exiting the first separating device, the steam produced in the heat recovery unit, and a gas stream, wherein said reacting produces heat and the first mixture product, wherein said gas stream comprises an oxygen supply selected from the group consisting of an oxygen-containing gas, steam, and mixtures thereof, and wherein the produced heat is recovered by converting the solid feedstock stream into the second mixture product in the reactor upper section.

25 **[0010]** In certain embodiments of the system, the second solid product stream exiting the first separating device, the steam exiting the heat recovery unit, and the gas stream of part (k) may be passed into the reactor lower section by one or more dispersion devices. The carbonaceous feedstock may be selected from coal, lignite, petroleum coke or mixtures thereof. The oxygen-containing gas may be air, oxygen-enriched air, oxygen or mixtures thereof. The heat recovery unit may be a radiant heat type boiler, water tube boiler, fire tube boiler or combinations thereof. The temperature of the second mixture product prior to entering into the first separating device is between 648°C (1200°F) and 1371°C (2500°F), but is preferably between 815°C (1500°F) and 1093°C (2000°F). The warm solid-gas mixture is preferably maintained at a temperature between 260°C (500°F) to 426°C (800°F). 30

BRIEF DESCRIPTION OF THE DRAWINGS

35 **[0011]** For a more detailed description of the embodiments, reference will now be made to the accompanying drawings, wherein:

40 FIG. 1 is a schematic representation of a system useful in and a pictorial process flow diagram for an example, which is not part of the present invention.

FIG. 2 is a schematic representation of a system useful in and a pictorial process flow diagram for an alternative embodiment of the present invention.

DETAILED DESCRIPTION

45 **[0012]** The following detailed description references the accompanying drawings which illustrate specific examples not according to the invention and embodiments in which the invention can be practiced. The embodiments are intended to describe aspects of the invention in sufficient detail to enable those skilled in the art to practice the invention. However, other embodiments can be utilized and changes can be made without departing from the scope of the present invention. Thus, the scope of the present invention is not limited to only the specific embodiments disclosed herein, but rather, the scope is defined only by the appended claims. 50

[0013] Referring to the example of FIG. 1 and the embodiment as shown in FIG. 2, various embodiments of the present invention provide a gasification reactor, indicated generally by reference numeral 10, that has a reactor lower section 30 and a reactor upper section 40. The first stage of the gasification process takes place in the reactor lower section 30 and the second stage of the gasification process takes place in the reactor upper section 40. The reactor lower section 30 defines the first stage reaction zone, and will alternatively be referred to as the first stage reaction zone. The reactor upper section 40 defines the second stage reaction zone, and will alternatively be referred to as the second stage reaction zone. 55

[0014] According to the example depicted in FIG. 1 and the embodiment as shown in FIG. 2, solid feedstock is pulverized (by methods that are known in the art, but outside the scope of this disclosure) before entering a feeding system 100 such as, but not limited to, a lock-hopper system. The pulverized solid stream comprising particulate carbonaceous material from the feeding system 100 is injected into the gasification reactor 10 upper section 40 through feeding device 80 and/or 80a, or additional feeding devices (not shown). The carbonaceous material then comes into contact with the hot syngas rising from the gasification reactor 10 lower section 30. The carbonaceous material is dried and a portion of it is gasified via pyrolysis reactions such as the carbon steam reaction ($C + H_2O \rightarrow CO + H_2$). Pyrolysis reactions are endothermic, thus, the temperature of the mixture of carbonaceous material and syngas decreases as the mixture travels upwards through the upper section 40. By the time the second mixture product comprising un-reacted solid particulates (e.g. char) and a second gaseous product stream (e.g. syngas) leaves the top of the upper section 40 of the gasifier 10, the second mixture product temperature drops to the range between 648°C (1200°F) and 1371°C (2500°F), but preferably to the range between 815°C (1500°F) and 1093°C (2000°F).

[0015] Further according to the example as shown in FIG. 1 and the embodiment as shown in FIG. 2, the second mixture product, comprising un-reacted solid particulates and a second gaseous product stream, exits the reactor upper section 40 and is sent to a first separating device 50. The first separating device 50 splits the second mixture product into a second solid product stream and second gaseous product stream, leaving only a small fraction of residual solid fines in the second gaseous product stream. The second solids product stream falls by gravitational force, and exits the first separating device 50 via an outlet 70. The second solids product stream is then recycled back to the reactor lower section 30 of the gasifier 10 through dispersion devices 60 and/or 60a. These devices mix the recycled solids with gaseous oxidant during addition of the solids and oxidant to the first stage of the reactor. The construction of such dispersion devices is commonly understood by those having average skill in the art.

[0016] Further according to the example as shown in FIG. 1 and the embodiment as shown in FIG. 2, the second solids product stream (primarily including char) reacts with oxygen in the presence of superheated steam in the lower section 30 (or first stage reaction zone) of the gasifier 10. The primary reactions in the first stage are $C + O_2 \rightarrow CO_2$ and $C + \frac{1}{2} O_2 \rightarrow CO$. These exothermic reactions raise the temperature of the gas in the first stage to between 1093°C (2000°F) and 1926°C (3500°F). The hot syngas produced in the reactor lower section 30 flows upward to the reactor upper section 40 where it comes into contact with the carbonaceous feedstock. The feedstock particles are dried and heated to an elevated temperature by the hot syngas, then the dry particles react with steam to generate CO and hydrogen. The primary reactions in the second stage are the carbon-steam reaction and $C + H_2O \rightarrow CO + H_2$ and the water-gas reaction $CO + H_2O \rightarrow CO_2 + H_2$. The carbon-steam reaction forms CO and H₂, thus, increasing the yield of these usable gases.

[0017] Again referring to the example as shown in FIG. 1 and the embodiment as shown in FIG. 2, the temperature of the first stage is higher than the ash melting point. Consequently, entrained ash particles agglomerate and become a viscous molten slag that flows down the sides of the gasifier to exit the reactor via the taphole 20 and enter the quench chamber. The slag is water-quenched and ultimately collected as a solid slag product. Typically, the steam added to the reactor lower section 30 is generated in the heat recovery unit 180. Water 170 is fed into the heat recovery unit 180, and is heated by the hot syngas exiting the upper section 40 of the gasifier 10. The produced steam is then routed to the lower section 30 of the gasifier 10 via dispersion devices 60 and/or 60a.

[0018] Further referring to FIG. 1, the second gaseous product stream exiting from the first separating device 50 comprises hydrogen, carbon monoxide, a small amount of methane, hydrogen sulfide, ammonia, nitrogen, carbon dioxide and small fraction of residual solid fines. After passing through the heat recovery unit 180, the cooled syngas is subsequently introduced into a particulate filtering device 110, whereby the residual solid fines and particulates are removed and recycled back to lower section 30 of the gasifier 10.

[0019] In the embodiment depicted in FIG. 2, solid feedstock is pulverized before entering a feeding system 100 such as, but not limited to, a lock-hopper system. The pulverized solid stream comprising particulate carbonaceous material from the feeding system is sent to a solid-gas mixer 160 where it contacts with warm syngas exiting the heat recovery unit 180. The function of the mixer 160 is to provide sufficient residence time to reduce feedstock moisture content, thereby producing a substantially dry feedstock. The temperature of the warm syngas from the heat recovery unit 180 is kept in a range between about 148°C (300°F) and 537°C (1000°F), but preferably in a range between about 260°C (500°F) and 426°C (800°F) to avoid formation of tars in the mixer 160. Tar formation is not desirable, as it contaminates the syngas and makes downstream gas and wastewater treatment more expensive.

[0020] Further referring to FIG. 2, the warm solid-gas mixture exiting mixer 160 is passed through a second separating device 150, which splits the warm solid-gas mixture into a second solid product stream and a second gaseous product stream, leaving only a small fraction of residual solid fines in the gas stream. In certain embodiments, the first separating device may comprise a cyclone or other commercially-available method for separating particles from a gas stream. The second solid product stream leaving the second separating device 150 is recycled back to the reactor upper section 40 of the gasifier 10 via dispersion devices 80 and/or 80a, or additional feeding devices (not shown). The second gaseous product stream leaving the second separating device 150 is next introduced into a particulate filtering device 110, whereby

residual solid fines and particulates are removed and recycled back to the upper section 40 of the gasifier 10 through feeding devices 80 and/or 80a (or additional feeding devices) as feedstock for the second stage reaction. The gaseous product exiting filtering device 110 comprises a raw syngas that is largely free of particulates. This raw syngas can then be further cleaned using processes that are outside the scope of this disclosure.

5 **[0021]** Further referring to FIG. 2, both the second solid product leaving the second separating device 150 and residual solid fines and particulates exiting from the particulate filtering device 110 then come into contact with the hot syngas rising from the lower section 30 of the gasifier 10. The carbonaceous material is de-volatilized and a portion of the solids is gasified via pyrolysis reactions in which H_2 and CO are produced. The un-reacted solids are essentially char and ash. The pyrolysis reactions that predominant in the upper section 40 of the gasifier 10 are highly endothermic. Thus, the temperature of the carbonaceous material and syngas mixture decreases as it travels upwards through the upper section 40. By the time the second mixture product, comprising the second solid product stream (e.g. char) and second gaseous product stream (e.g. syngas) leave the top of the upper section 40 of the gasifier 10, the temperature of the second mixture product is in the range between 648°C (1200°F) and 1371°C (2500°F), but more preferably in the range between 815°C (1500°F) and 1093°C (2000°F).

10 **[0022]** Further referring to FIG. 2, the second mixture product comprising the second solid product stream and second gaseous product stream exiting the reactor upper section 40 is sent to a first separating device 50, which splits the mixture into a second solid product stream and second gaseous product stream, leaving only a small fraction of residual solid fines in the second gaseous product stream. The second solids product stream exiting separating device 50 is recycled back to the reactor lower section 30 of the gasifier 10 through dispersion devices 60 and/or 60a as feedstock for the first stage reaction.

20 **[0023]** Further according to the embodiment as shown in FIG. 2, the second solids product stream (primarily including char) reacts with oxygen in the presence of superheated steam in the first stage in the lower section 30 of the gasifier 10. The primary reactions occurring in the first stage include $C + O_2 \rightarrow CO_2$ and $C + \frac{1}{2} O_2 \rightarrow CO$ which are both highly exothermic. As a result, the temperature within the first stage is maintained in a range between 1093°C (2000°F) and 1926°C (3500°F). The heat produced by the first stage reaction zone 30 and carried upward by the gas stream is used for the second stage pyrolysis reactions that predominate in the unfired reactor upper section 40, including vaporization of the moisture carried in by the feed, the carbon-steam reaction and the water-gas reaction between the CO and H_2O .

25 **[0024]** Again referring to the embodiment as shown in FIG. 2, the temperature of the first stage is higher than the ash melting point. Consequently, entrained ash particles agglomerate and become a viscous molten slag that flows down the sides of the gasifier to exit the reactor via the taphole 20 and enter the quench chamber. The slag is water-quenched and ultimately collected as a solid slag product. The steam added to the reactor lower section 30 may be generated from the heat recovery unit 180 using the heat from the hot syngas exiting from the second stage (upper section) of the gasifier 10.

30 **[0025]** Further referring to FIG. 2, the second gaseous product stream exiting from first separating device 50 comprises hydrogen, carbon monoxide, a small amount of methane, hydrogen sulfide, ammonia, nitrogen, carbon dioxide and small fraction of residual solid fines. After passing through the heat recovery unit 180, the warm syngas is sent to a mixer 160 where it comes into contact with pulverized solid feedstock, thereby forming a warm solid-gas mixture that serves to dry the feedstock. The temperature of the warm solid-gas mixture in mixer 160 is kept in a range between about 148°C (300°F) and 537°C (1000°F), but preferably between about 260°C (500°F) and 426°C (800°F) to minimize formation of tar. The warm solid-gas mixture exiting mixer 160 is next introduced into a particulate filtering device 110, whereby the residual solid fines and particulates are removed and recycled back to the upper section 40 of the gasifier 10, as previously described above.

35 **[0026]** In certain embodiments as shown in FIG. 2, and as illustrated in the example of FIG. 1, the recycled char, a stream of an oxygen-containing gas, and steam enter the gasification reactor 10 lower section 30 through dispersion devices 60 and/or 60a, which are located at either end of the horizontally extended portions of the lower section 30. More than two dispersion devices can be used, for example, four, arranged 90 degrees apart. The sets of dispersion devices can also be on different levels and need not be on the same plane.

40 **[0027]** Again referring to the example depicted in FIG. 1 and the embodiment depicted in FIG. 2, the unfired reactor upper section 40 connects directly to the top of the fired reactor lower section 30 so that the hot reaction products are conveyed directly from the reactor lower section 30 to the reactor upper section 40. This minimizes heat losses in the gaseous reaction products and entrained solids, thereby increasing process efficiency.

45 **[0028]** Further referring to the example depicted in FIG. 1 and the embodiment depicted in FIG. 2, the dispersion devices 60 and/or 60a provide an atomized feed of the particulate solids such as char. The dispersion devices may be of the type having a central tube for the solids and an annular space surrounding the central tube containing the atomizing gas which opens to a common mixing zone internally or externally. Further, the feeding device 80 and/or 80a of the unfired reactor upper section 40 may also be similar to the dispersion devices described hereinabove. Dispersion devices 60 and/or 60a, or feeding devices 80 and/or 80a can be as are conventionally known to those skilled in the art.

50 **[0029]** The materials used to construct the gasification reactor 10 are not critical. Preferably, but not necessarily, the

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reactor walls are steel and are lined with an insulating castable or ceramic fiber or refractory brick, such as a high chrome-containing brick in the reactor lower section 30 and a dense medium, such as used in blast furnaces and non-slugging applications in the reactor upper section 40, in order to reduce heat loss and to protect the vessel from high temperature and corrosive molten slag as well as to provide for better temperature control, all of which are commercially available from several sources. Use of this type of system provides the high recovery of heat values from the carbonaceous solids used in the process. Optionally and alternatively, the walls may be unlined by providing a "cold wall" system for fired reactor lower section 30 and, optionally, unfired upper section 40. The term "cold wall", as used herein, means that the walls are cooled by a cooling jacket with a cooling medium, as is known conventionally in the art for prior art coal gasification systems. In such a system, the slag freezes on the cooled interior wall and thereby protects the metal walls of the cooling jacket against heat degradation.

[0030] The physical conditions of the reaction in the first stage of the process in the reactor lower section 30 are controlled and maintained to assure rapid gasification of the char at temperatures exceeding the melting point of ash to produce a molten slag from the melted ash having a viscosity not greater than approximately 25 Pa.s (250 poises). This slag drains from the reactor through the taphole 20, and is further processed in units outside the scope of this document.

[0031] The physical conditions of the reaction in the second stage of the gasification process in the reactor upper section 40 are controlled to assure rapid gasification and heating of the coal above its range of plasticity. The temperature of the reactor lower section 30 is maintained in a range between 815°C (1500°F) and 1926°C (3500°F), preferably in a range between 1093°C (2000°F) and 1760°C (3200°F) and most preferably in a range between 1204°C (2200°F) and 1648°C (3000°F). Pressures inside both the reactor upper section 40 and lower section 30 of the gasifier 10 are maintained at atmospheric pressure or higher.

[0032] As used herein, the term "oxygen-containing gas" that is fed to the reactor lower section 30 is defined as any gas containing at least 20 percent oxygen. Preferred oxygen-containing gases include oxygen, air, and oxygen-enriched air.

[0033] Any particulate carbonaceous material can be utilized as feedstock for the embodiments described herein. Preferably, however, the particulate carbonaceous material is coal, which without limitation includes lignite, bituminous coal, sub-bituminous coal, and any combinations thereof. Additional carbonaceous materials are coke derived from coal, coal char, coal liquefaction residue, particulate carbon, petroleum coke, carbonaceous solids derived from oil shale, tar sands, pitch, biomass, concentrated sewer sludge, bits of garbage, rubber and mixtures thereof. The foregoing exemplified materials can be in the form of comminuted solids.

[0034] When coal or petroleum coke is the feedstock, it can be pulverized before addition to the reactor upper section. In general, any finely-divided carbonaceous material may be used, and any of the known methods of reducing the particle size of particulate solids may be employed. Examples of such methods include the use of ball, rod and hammer mills. While particle size is not critical, finely divided carbon particles are preferred. Powdered coal used as fuel in coal-fed power plants is typical. Such coal has a particle size distribution such that 90% (by weight) of the coal passes through a 200 mesh sieve. A coarser size of 100 mesh average particle size can also be used for more reactive materials, provided that a stable and non-settling slurry can be prepared.

[0035] As used herein, the term "char" refers to unburned carbon and ash particles that remain entrained within a gasification system after production of the various products.

[0036] As used herein, the term "substantially dry" means lacking in significant moisture content, but is not necessarily synonymous with absolute dryness.

EXAMPLE

[0037] Table 1 demonstrates the performance of the two-stage dry feed process and system as described herein. A computer simulation of the process as described herein was performed, based on Illinois 6 coal, with the gasifier pressure set at 3.60 MPa (523 psia), and the gas temperature leaving the second stage of the gasifier was set at 1032°C (1890°F). With the process parameters as listed in the Table 1, the calculated cold gas efficiency for the process was 84.6%. This calculated cold gas efficiency is 10% higher than that achieved by the current slurry-fed E-Gas™ gasification process (Conoco Phillips Co.), while oxygen consumption is calculated to be reduced by 15% versus the E-Gas™ process.

TABLE 1

Oxygen rate (kg/hr (lb/hr))	6.98E+04 (1.54E+05)
Coal HHV (dry), (kJ/kg (btu/lb))	29075 (12,500)
Coal (dry) flow rate (kg/hr (lb/hr))	1.01E+05 (2.23E+05)
Total HHV of Coal, (MJ/hr (btu/hr))	2.94E+12 (2.79E+09)

(continued)

Net syngas yield:	
H ₂ (kg/hr (lb/hr))	7.62E+03 (1.68E+04)
CH ₄ (kg/hr (lb/hr))	1.10E+02 (2.43E+02)
CO (kg/hr (lb/hr))	1.38E+5 (3.06E+05)
Total HHV of syngas (MJ/hr (btu/hr)) (H ₂ , CO, and CH ₄)	2.48E+12 (2.36E+09)
Oxygen/coal (dry), (wt/wt)	0.69
Cold syngas efficiency (HHV)	84.6%

[0038] The scope of protection sought is not intended to be limited by the description or examples set out above, but only by the claims that follow.

Claims

1. A process for gasifying a carbonaceous material, comprising the steps of:

- (a) providing a gasification reactor (10) comprising a reactor upper section (40) and a reactor lower section (30);
 (b) introducing a substantially dry solid carbonaceous feedstock stream, optionally selected from the group consisting of coal, lignite, petroleum coke and mixtures thereof, into the reactor upper section (40) and reacting therein with a first mixture product from the reactor lower section (30), thereby forming a second mixture product, wherein the first mixture product comprises syngas, and wherein the second mixture product comprises a second solid product and a second gaseous product;
 (c) passing the second mixture product to a first separating device (50), wherein the second solid product is separated from the second gaseous product;
 (d) passing the separated second solid product into the reactor lower section (30);
 (e) passing the second gaseous product stream exiting the first separating device (50) through a heat recovery unit (180), thereby lowering the temperature of the second gaseous product stream to provide a cooled second gaseous product and creating steam;
 (f) passing the steam produced in step (e) into the reactor lower section (30);
 (g) pulverizing a solid carbonaceous feedstock;
 (h) mixing the pulverized solid carbonaceous feedstock with the cooled second gaseous product of step (e) by means of a solid-gas mixer (160) to form a warm solid-gas mixture, wherein said warm solid-gas mixture is maintained at a temperature in a range of 148°C (300°F) to 537°C (1000°F);
 (i) separating the warm solid-gas mixture in a second separating device (150) to produce the substantially dry solid carbonaceous feedstock of step (b) and a separated second gaseous product;
 (j) passing the separated second gaseous product exiting the second separating device (150) to a particulate filtering device (110), wherein a stream of residual solids, fines and particulates is removed and passed into the reactor upper section (40); and
 (k) combining a gas stream, the separated second solid product stream of step (d) and the steam produced in the heat recovery unit (180) of step (e) in the reactor lower section (30) and reacting them, thereby evolving heat and forming the first mixture product comprising syngas of step (b), wherein the gas stream comprises an oxygen supply selected from the group consisting of an oxygen-containing gas, steam, and mixtures thereof.

2. The process of claim 1, wherein said substantially dry solid carbonaceous feedstock stream and said residual solids, fines and particulates are passed into said reactor upper section (40) by one or more feeding devices (80, 80a).

3. The process of claim 1, wherein the gas stream of part (k), the second solid product exiting the first separating device (50), and the steam produced inside the heat recovery unit (180) are passed into the reactor lower section (30) by one or more dispersion devices (60, 60a).

4. The process of claim 1,

wherein the oxygen-containing gas is selected from the group consisting of air, oxygen-enriched air, oxygen and mixtures thereof,
 wherein the heat recovery unit (180) is selected from the group consisting of a radiant heat type boiler, water tube boiler, fire tube boiler and combinations thereof.

- 5
5. The process of claim 1, wherein the temperature of the second mixture product prior to entering into said first separating device is between 648°C (1200°F) and 1371°C (2500°F), optionally between 816°C (1500°F) and 1093°C (2000°F).
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6. The process of claim 1, wherein the warm solid-gas mixture is maintained at a temperature in a range of 260°C (500°F) to 426°C (800°F).
7. The process of claim 1, wherein the first and second separating devices (50, 150) each comprise a cyclone.
- 15
8. A system for gasification of a carbonaceous material, comprising:
- (a) a reactor upper section (40) for reacting:
- 20
- i) a substantially dry solid carbonaceous feedstock, optionally selected from the group consisting of coal, lignite, petroleum coke and mixtures thereof,
- ii) recycled residual solids, fines and particulates, and
- iii) a first mixture product from a reactor lower section (30) to produce a second mixture product, wherein the first mixture product comprises syngas, and the second mixture product comprises a second solid product stream and a second gaseous product stream;
- 25
- (b) a first separating device (50) for separating the second solid product stream from the second gaseous product stream,
 wherein the second solid product is introduced into the reactor lower section (30);
- (c) a heat recovery unit (180) for cooling the temperature of the second gaseous product stream to provide a cooled second gaseous product and producing steam,
 wherein said steam is passed into said lower reactor section;
- 30
- (d) a solid-gas mixer (160) for mixing a pulverized solid carbonaceous feedstock with the cooled second gaseous product of part (c) to form a warm solid-gas mixture, wherein the solid-gas mixer (160) is configured for maintaining the warm solid-gas mixture at a temperature in a range of 148°C (300°F) to 537°C (1000°F);
- 35
- (e) a second separating device (150) for separating the warm solid-gas mixture to produce the substantially dry solid carbonaceous feedstock of part (a) and a separated second gaseous product;
- (f) a particulate filtering device (110) for separating the residual solids, fines and particulates from the separated second gaseous product stream exiting the second separating device (150),
 wherein the separated residual solids, fines and particulates are passed into the reactor upper section (40);
- 40
- (g) a reactor lower section (30) for reacting a mixture comprising:
- i) the separated second solid product exiting the first separating device (50);
- ii) the steam produced in the heat recovery unit (180);
- iii) a gas stream;
- 45
- wherein said reacting produces heat and said first mixture product of part (a),
 wherein the gas stream comprises an oxygen supply selected from the group consisting of oxygen-containing gas, steam, and mixtures thereof, and wh
 erein the heat produced in the reactor lower section (30) is recovered by converting the solid feedstock stream
 into the second mixture product in the reactor upper section (40).
- 50
9. The system of claim 8, wherein the second solid product stream exiting the first separating device (50), the steam exiting the heat recovery unit (180), and the gas stream of part (k) are passed into the reactor lower section (30) by one or more dispersion devices (60, 60a).
- 55
10. The system of claim 8,

wherein the oxygen-containing gas is selected from the group consisting of air, oxygen-enriched air, oxygen

and mixtures thereof,

wherein the heat recovery unit (180) is selected from the group consisting of a radiant heat type boiler, water tube boiler, fire tube boiler and combinations thereof.

5 11. The system of claim 8, wherein the solid-gas mixer (160) is configured for maintaining the warm solid-gas mixture at a temperature between 260°C (500°F) and 426°C (800°F).

12. The system of claim 8, wherein the first and second separating devices (50, 150) each comprise a cyclone.

10

Patentansprüche

1. Verfahren zur Vergasung eines kohlenstoffhaltigen Materials, umfassend die Schritte:

15 (a) Bereitstellen eines Vergasungsreaktors (10), der einen oberen Reaktorabschnitt (40) und einen unteren Reaktorabschnitt (30) umfasst;

(b) Einführen eines Stroms von im Wesentlichen trockenem festen kohlenstoffhaltigen Einsatzmaterial, das gegebenenfalls aus der Gruppe ausgewählt ist, die aus Steinkohle, Braunkohle, Petrolkoks und Gemischen davon besteht, in den oberen Reaktorabschnitt (40) und darin Umsetzen mit einem ersten Mischungsprodukt aus dem unteren Reaktorabschnitt (30) unter Bildung eines zweiten Mischungsprodukts, wobei das erste Mischungsprodukt Synthesegas umfasst, und

wobei das zweite Mischungsprodukt ein zweites festes Produkt und ein zweites gasförmiges Produkt umfasst; (c) Leiten des zweiten Mischungsprodukts in eine erste Trennvorrichtung (50), wobei das zweite feste Produkt von dem zweiten gasförmigen Produkt getrennt wird;

25 (d) Leiten des abgetrennten zweiten festen Produkts in den unteren Reaktorabschnitt (30);

(e) Leiten des zweiten gasförmigen Produktstroms, der aus der ersten Trennvorrichtung (50) austritt, durch eine Wärmerückgewinnungseinheit (180), wodurch die Temperatur des zweiten gasförmigen Produktstroms gesenkt wird, wobei man ein abgekühltes zweites gasförmiges Produkt erhält und Dampf erzeugt;

(f) Leiten des in Schritt (e) erzeugten Dampfes in den unteren Reaktorabschnitt (30);

30 (g) Pulverisieren eines festen kohlenstoffhaltigen Einsatzmaterials;

(h) Mischen des pulverisierten festen kohlenstoffhaltigen Einsatzmaterials mit dem abgekühlten zweiten gasförmigen Produkt aus Schritt (e) mittels eines Feststoff-Gas-Mischers (160) unter Bildung eines warmen Feststoff-Gas-Gemischs, wobei das warme Feststoff-Gas-Gemisch auf einer Temperatur im Bereich von 148 °C (300 °F) bis 537 °C (1000 °F) gehalten wird;

35 (i) Trennen des warmen Feststoff-Gas-Gemischs in einer zweiten Trennvorrichtung (150), wobei das im Wesentlichen trockene feste kohlenstoffhaltige Einsatzmaterial von Schritt (b) und ein abgetrenntes zweites gasförmiges Produkt entstehen;

(j) Leiten des abgetrennten zweiten gasförmigen Produkts, das aus der zweiten Trennvorrichtung (150) austritt, in eine partikuläre Filtervorrichtung (110),

40 wobei ein Strom von restlichen Feststoffen, Feinstoffen und partikulären Stoffen entfernt und in den oberen Reaktorabschnitt (40) geleitet wird; und

(k) Kombinieren eines Gasstroms, des abgetrennten zweiten festen Produktstroms von Schritt (d) und des in der Wärmerückgewinnungseinheit (180) von Schritt (e) erzeugten Dampfes in dem unteren Reaktorabschnitt (30) und Umsetzen derselben, wodurch sich Wärme entwickelt und das erste Mischungsprodukt entsteht, das Synthesegas aus Schritt (b) umfasst,

45 wobei der Gasstrom eine Sauerstoffquelle umfasst, die aus der Gruppe ausgewählt ist, die aus sauerstoffhaltigem Gas, Dampf und Gemischen davon besteht.

2. Verfahren gemäß Anspruch 1, wobei der Strom des im Wesentlichen trockenen festen kohlenstoffhaltigen Einsatzmaterials und die restlichen Feststoffe, Feinstoffe und partikulären Stoffe durch eine oder mehrere Zuführungsvorrichtungen (80, 80a) in den oberen Reaktorabschnitt (40) geleitet werden.

3. Verfahren gemäß Anspruch 1, wobei der Gasstrom von Teil (k), das zweite feste Produkt, das aus der ersten Trennvorrichtung (50) austritt, und der in der Wärmerückgewinnungseinheit (180) erzeugte Dampf durch eine oder mehrere Dispersionsvorrichtungen (60, 60a) in den unteren Reaktorabschnitt (30) geleitet werden.

4. Verfahren gemäß Anspruch 1, wobei das sauerstoffhaltige Gas aus der Gruppe ausgewählt ist, die aus Luft, mit Sauerstoff angereicherter Luft,

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Sauerstoff und Gemischen davon besteht,

wobei die Wärmerückgewinnungseinheit (180) aus der Gruppe ausgewählt ist, die aus einem Radiatorkessel, Wasserröhrenkessel, Flammröhrenkessel und Kombinationen davon besteht.

- 5 5. Verfahren gemäß Anspruch 1, wobei die Temperatur des zweiten Mischungsprodukts vor dem Eintritt in die erste Trennvorrichtung zwischen 648 °C (1200 °F) und 1371 °C (2500 °F), gegebenenfalls zwischen 816 °C (1500 °F) und 1093 °C (2000 °F), liegt.
- 10 6. Verfahren gemäß Anspruch 1, wobei das warme Feststoff-Gas-Gemisch auf einer Temperatur im Bereich von 260 °C (500 °F) bis 426 °C (800 °F) gehalten wird.
7. Verfahren gemäß Anspruch 1, wobei die erste und die zweite Trennvorrichtung (50, 150) jeweils einen Fliehkraftabscheider umfassen.
- 15 8. System zur Vergasung eines kohlenstoffhaltigen Materials, umfassend:
- (a) einen oberen Reaktorabschnitt (40) zum Umsetzen von:
- 20 i) einem im Wesentlichen trockenen festen kohlenstoffhaltigen Einsatzmaterial, das gegebenenfalls aus der Gruppe ausgewählt ist, die aus Steinkohle, Braunkohle, Petrolkoks und Gemischen davon besteht,
- ii) im Kreislauf zurückgeführten restlichen Feststoffen, Feinstoffen und partikulären Stoffen, und
- iii) einem ersten Mischungsprodukt aus einem unteren Reaktorabschnitt (30) unter Bildung eines zweiten Mischungsprodukts,
- 25 wobei das erste Mischungsprodukt Synthesegas umfasst und das zweite Mischungsprodukt einen Strom eines zweiten festen Produkts und einen Strom eines zweiten gasförmigen Produkts umfasst;
- (b) eine erste Trennvorrichtung (50) zum Abtrennen des Stroms des zweiten festen Produkts von dem Strom des zweiten gasförmigen Produkts,
- 30 wobei das zweite feste Produkt in den unteren Reaktorabschnitt (30) eingeführt wird;
- (c) eine Wärmerückgewinnungseinheit (180) zum Senken der Temperatur des zweiten gasförmigen Produktstroms, wobei man ein abgekühltes zweites gasförmiges Produkt erhält und Dampf erzeugt, wobei der Dampf in den unteren Reaktorabschnitt geleitet wird;
- (d) einen Feststoff-Gas-Mischer (160) zum Mischen eines pulverisierten festen kohlenstoffhaltigen Einsatzmaterials mit dem abgekühlten zweiten gasförmigen Produkt aus Teil (c) unter Bildung eines warmen Feststoff-Gas-Gemischs, wobei der Feststoff-Gas-Mischer (160) so konfiguriert ist, dass er das warme Feststoff-Gas-Gemisch auf einer Temperatur im Bereich von 148 °C (300 °F) bis 537 °C (1000 °F) halten kann;
- 35 (e) eine zweite Trennvorrichtung (150) zum Trennen des warmen Feststoff-Gas-Gemischs, wobei das im Wesentlichen trockene feste kohlenstoffhaltige Einsatzmaterial von Teil (a) und ein abgetrenntes zweites gasförmiges Produkt entstehen;
- 40 (f) eine partikuläre Filtervorrichtung (110) zum Abtrennen der restlichen Feststoffe, Feinstoffe und partikulären Stoffe von dem Strom des abgetrennten zweiten gasförmigen Produkts, das aus der zweiten Trennvorrichtung (150) austritt, wobei die restlichen Feststoffe, Feinstoffe und partikulären Stoffe in den oberen Reaktorabschnitt (40) geleitet werden;
- 45 (g) einen unteren Reaktorabschnitt (30) zum Umsetzen eines Gemischs, umfassend:
- i) das abgetrennte zweite feste Produkt, das aus der ersten Trennvorrichtung (50) austritt;
- 50 ii) den in der Wärmerückgewinnungseinheit (180) erzeugten Dampf;
- iii) einen Gasstrom;
- wobei das Umsetzen Wärme und das erste Mischungsprodukt von Teil (a) erzeugt;
- wobei der Gasstrom eine Sauerstoffquelle umfasst, die aus der Gruppe ausgewählt ist, die aus sauerstoffhaltigem Gas, Dampf und Gemischen davon besteht, und
- wobei die in dem unteren Reaktorabschnitt (30) erzeugte Wärme zurückgewonnen wird, indem man den Strom des festen Einsatzmaterials im oberen Reaktorabschnitt (40) in das zweite Mischungsprodukt umwandelt.
- 55 9. System gemäß Anspruch 8, wobei der zweite feste Produktstrom, der aus der ersten Trennvorrichtung (50) austritt, der Dampf, der aus der Wärmerückgewinnungseinheit (180) austritt, und der Gasstrom von Teil (k) durch eine oder

mehrere Dispersionsvorrichtungen (60, 60a) in den unteren Reaktorabschnitt (30) geleitet werden.

10. System gemäß Anspruch 8,
wobei das sauerstoffhaltige Gas aus der Gruppe ausgewählt ist, die aus Luft, mit Sauerstoff angereicherter Luft,
Sauerstoff und Gemischen davon besteht,
wobei die Wärmerückgewinnungseinheit (180) aus der Gruppe ausgewählt ist, die aus einem Radiatorkessel, Wasserrohrkessel, Flammrohrkessel und Kombinationen davon besteht.
11. System gemäß Anspruch 8, wobei der Feststoff-Gas-Mischer (160) so konfiguriert ist, dass er das warme Feststoff-Gas-Gemisch auf einer Temperatur im Bereich von 260 °C (500 °F) bis 426 °C (800 °F) halten kann.
12. System gemäß Anspruch 8, wobei die erste und die zweite Trennvorrichtung (50, 150) jeweils einen Fliehkraftabscheider umfassen.

Revendications

1. Procédé de gazéification d'une matière carbonée, comprenant les étapes consistant à:

(a) fournir un réacteur de gazéification (10) comprenant une section supérieure de réacteur (40) et une section inférieure de réacteur (30) ;

(b) introduire un flux de charge d'alimentation carbonée solide substantiellement sèche, optionnellement choisie dans le groupe constitué par le charbon, le lignite, le coke de pétrole et leurs mélanges, dans la section supérieure de réacteur (40) et l'y faire réagir avec un premier produit de mélange provenant de la section inférieure de réacteur (30), formant ainsi un deuxième produit de mélange, dans lequel le premier produit de mélange comprend du gaz de synthèse, et dans lequel le deuxième produit de mélange comprend un deuxième produit solide et un deuxième produit gazeux ;

(c) faire passer le deuxième produit de mélange dans un premier dispositif de séparation (50), dans lequel le deuxième produit solide est séparé du deuxième produit gazeux ;

(d) faire passer le deuxième produit solide séparé dans la section inférieure de réacteur (30) ;

(e) faire passer le deuxième flux de produit gazeux sortant du premier dispositif de séparation (50) à travers une unité de récupération de chaleur (180), ce qui permet d'abaisser la température du deuxième flux de produit gazeux pour obtenir un deuxième produit gazeux refroidi et de créer de la vapeur ;

(f) faire passer la vapeur produite à l'étape (e) dans la section inférieure de réacteur (30) ;

(g) pulvériser une charge d'alimentation carbonée solide ;

(h) mélanger la charge d'alimentation carbonée solide pulvérisée avec le deuxième produit gazeux refroidi de l'étape (e) au moyen d'un mélangeur solide-gaz (160) pour former un mélange solide-gaz chaud, dans lequel ledit mélange solide-gaz chaud est maintenu à une température comprise entre 148°C (300°F) et 537°C (1000°F) ;

(i) séparer le mélange solide-gaz chaud dans un deuxième dispositif de séparation (150) pour produire la charge d'alimentation carbonée solide substantiellement sèche de l'étape (b) et un deuxième produit gazeux séparé ;

(j) faire passer le deuxième produit gazeux séparé sortant du deuxième dispositif de séparation (150) vers un dispositif de filtrage de particules (110),

dans lequel un flux de solides résiduels, de fines et de particules est retiré et passé dans la section supérieure de réacteur (40) ; et

(k) combiner un flux de gaz, le deuxième flux de produit solide séparé de l'étape (d) et la vapeur produite dans l'unité de récupération de chaleur (180) de l'étape (e) dans la section inférieure de réacteur (30) et les faire réagir, ce qui fait évoluer la chaleur et forme le premier produit de mélange comprenant le gaz de synthèse de l'étape (b),

dans lequel le flux gazeux comprend une alimentation en oxygène choisie dans le groupe constitué par un gaz contenant de l'oxygène, de la vapeur et des mélanges de ceux-ci.

2. Procédé selon la revendication 1,

dans lequel ledit flux de charge d'alimentation carbonée solide substantiellement sèche et lesdits solides résiduels, fines et particules sont passés dans ladite section supérieure de réacteur (40) par un ou plusieurs dispositifs d'alimentation (80, 80a).

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3. Procédé selon la revendication 1,
dans lequel le flux gazeux de la partie (k), le deuxième produit solide sortant du premier dispositif de séparation (50) et la vapeur produite à l'intérieur de l'unité de récupération de chaleur (180) sont passés dans la section inférieure de réacteur (30) par un ou plusieurs dispositifs de dispersion (60, 60a).
- 5
4. Procédé selon la revendication 1,
dans lequel le gaz contenant de l'oxygène est choisi dans le groupe constitué par l'air, l'air enrichi en oxygène, l'oxygène et leurs mélanges,
10 dans lequel l'unité de récupération de chaleur (180) est choisie dans le groupe constitué par une chaudière de type à chaleur radiante, une chaudière à tubes d'eau, une chaudière à tubes de fumée et des combinaisons de celles-ci.
5. Procédé selon la revendication 1,
15 dans lequel la température du deuxième produit de mélange avant son entrée dans ledit premier dispositif de séparation est comprise entre 648°C (1200°F) et 1371°C (2500°F), optionnellement entre 816°C (1500°F) et 1093°C (2000°F).
6. Procédé selon la revendication 1,
20 dans lequel le mélange solide-gaz chaud est maintenu à une température comprise entre 260°C (500°F) et 426°C (800°F).
7. Procédé selon la revendication 1,
25 dans lequel les premier et deuxième dispositifs de séparation (50, 150) comprennent chacun un cyclone.
8. Système de gazéification d'un matériau carboné, comprenant :
- (a) une section supérieure de réacteur (40) pour la réaction :
- 30 i) d'une charge d'alimentation carbonée solide substantiellement sèche, optionnellement choisie dans le groupe constitué par le charbon, le lignite, le coke de pétrole et leurs mélanges,
ii) de solides résiduels, fines et particules recyclés, et
iii) d'un premier produit de mélange provenant d'une section inférieure de réacteur (30) pour produire un deuxième produit de mélange,
35 dans lequel le premier produit de mélange comprend du gaz de synthèse, et le deuxième produit de mélange comprend un deuxième flux de produit solide et un deuxième flux de produit gazeux ;
- (b) un premier dispositif de séparation (50) pour séparer le deuxième flux de produit solide du deuxième flux de produit gazeux,
40 dans lequel le deuxième produit solide est introduit dans la section inférieure de réacteur (30) ;
(c) une unité de récupération de chaleur (180) pour abaisser la température du deuxième flux de produit gazeux afin de fournir un deuxième produit gazeux refroidi et produire de la vapeur,
dans lequel ladite vapeur est passée dans ladite section inférieure de réacteur ;
(d) un mélangeur solide-gaz (160) pour mélanger une charge d'alimentation carbonée solide pulvérisée avec
45 le deuxième produit gazeux refroidi de la partie (c) pour former un mélange solide-gaz chaud, dans lequel le mélangeur solide-gaz (160) est configuré pour maintenir le mélange solide-gaz chaud à une température comprise entre 148°C (300°F) et 537°C (1000°F) ;
(e) un deuxième dispositif de séparation (150) pour séparer le mélange solide-gaz chaud afin de produire la charge d'alimentation carbonée solide substantiellement sèche de la partie (a) et un deuxième produit gazeux
50 séparé ;
(f) un dispositif de filtrage de particules (110) pour séparer les solides résiduels, les fines et les particules du deuxième flux de produit gazeux séparé à la sortie du deuxième dispositif de séparation (150),
dans lequel les solides résiduels, les fines et les particules séparés sont passés dans la section supérieure de réacteur (40) ;
55 (g) une section inférieure de réacteur (30) pour la réaction d'un mélange comprenant:
i) le deuxième produit solide séparé sortant du premier dispositif de séparation (50) ;
ii) la vapeur produite dans l'unité de récupération de chaleur (180) ;

iii) un flux de gaz ;

dans lequel ladite réaction produit de la chaleur et ledit premier produit de mélange de la partie (a),
dans lequel le flux gazeux comprend une alimentation en oxygène choisie dans le groupe constitué par un gaz
contenant de l'oxygène, de la vapeur et des mélanges de ceux-ci, et
dans lequel la chaleur produite dans la section inférieure de réacteur (30) est récupérée en convertissant le
flux de charge d'alimentation solide en un deuxième produit de mélange dans la section supérieure de réacteur
(40).

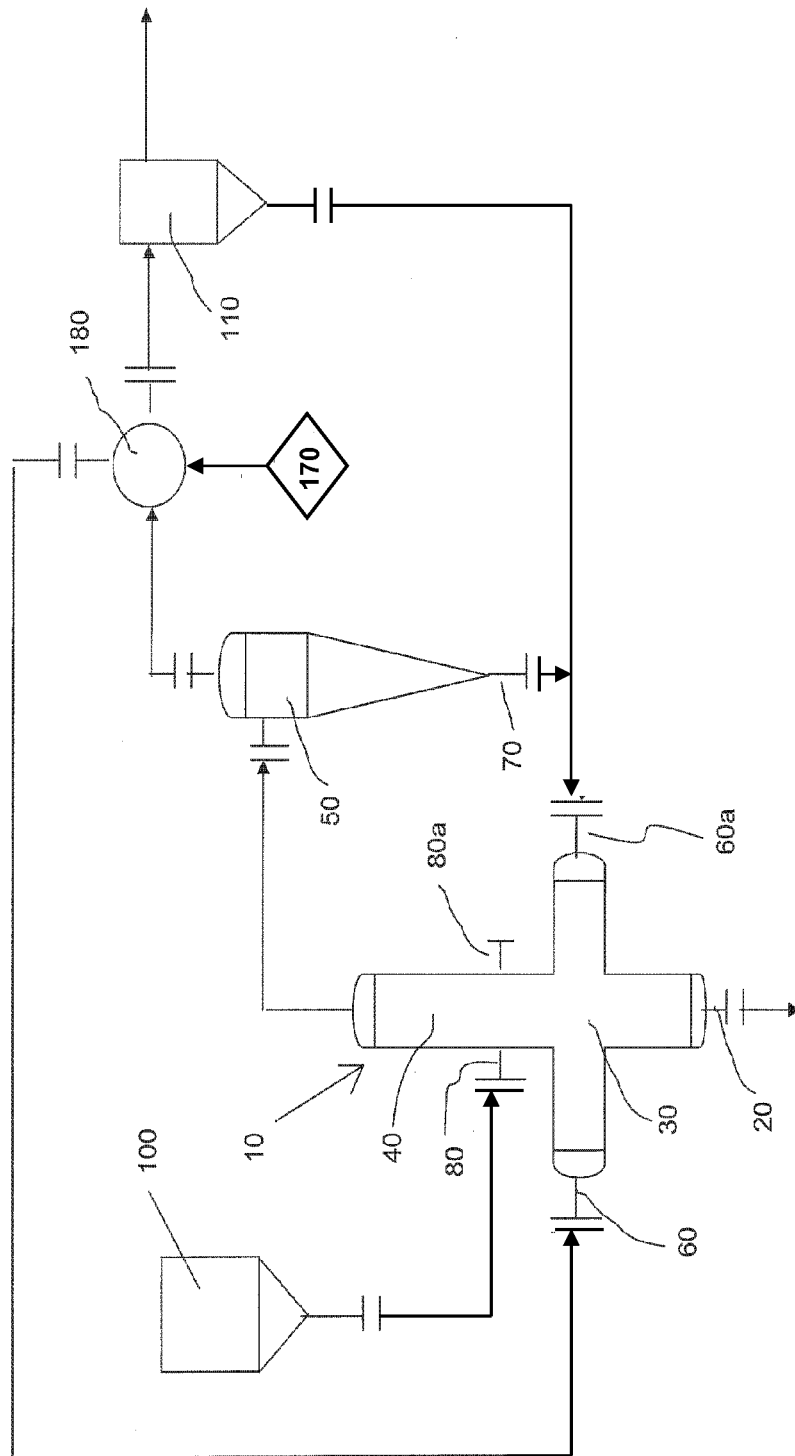
9. Système selon la revendication 8,
dans lequel le deuxième flux de produit solide sortant du premier dispositif de séparation (50), la vapeur sortant de
l'unité de récupération de chaleur (180) et le flux de gaz de la partie (k) sont passés dans la section inférieure de
réacteur (30) par un ou plusieurs dispositifs de dispersion (60, 60a).

10. Système selon la revendication 8,
dans lequel le gaz contenant de l'oxygène est choisi dans le groupe constitué par l'air, l'air enrichi en oxygène,
l'oxygène et leurs mélanges,
dans lequel l'unité de récupération de chaleur (180) est choisie dans le groupe constitué par une chaudière de
type à chaleur radiante, une chaudière à tubes d'eau, une chaudière à tubes de fumée et des combinaisons
de celles-ci.

11. Système selon la revendication 8,
dans lequel le mélangeur solide-gaz (160) est configuré pour maintenir le mélange solide-gaz chaud à une tempé-
rature comprise entre 260°C (500°F) et 426°C (800°F).

12. Système selon la revendication 8,
dans lequel les premier et deuxième dispositifs de séparation (50, 150) comprennent chacun un cyclone.

FIG. 1:



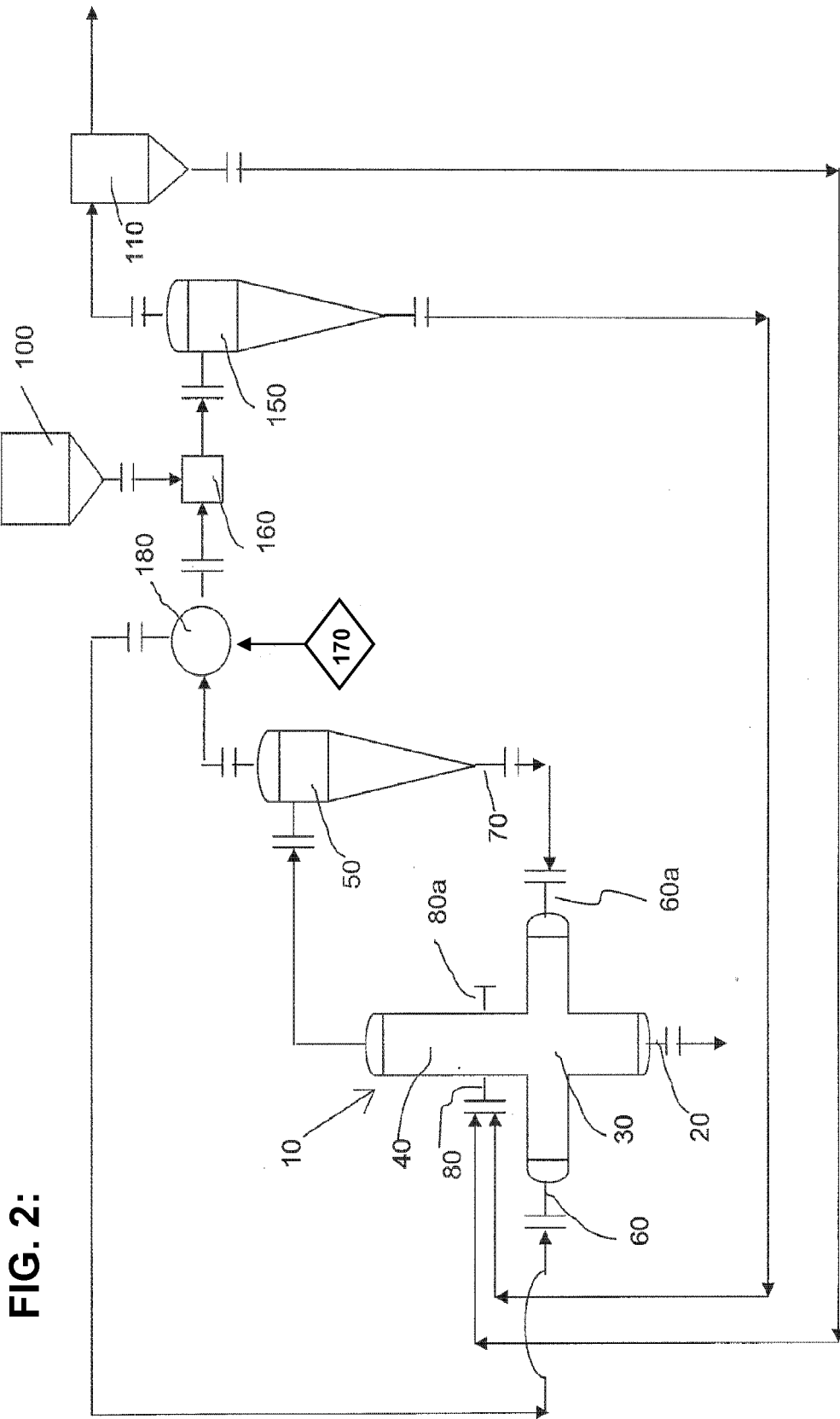


FIG. 2:

REFERENCES CITED IN THE DESCRIPTION

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